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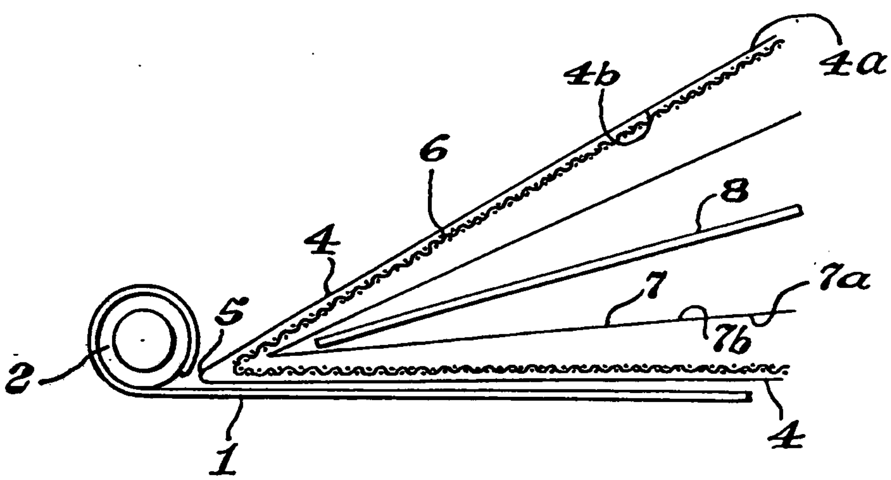


**(57) Abstract**

A spiral wound membrane separation device providing three fluid flow channels separated by semi-permeable membranes (4, 7) is disclosed. When enclosed in a containment vessel each fluid flow channel may be connected by ports to external fluid connections. Both closed ended flow into, and flow through, each channel is possible. A method of separating a selected segment of components of a fluid mixture is disclosed. The module is particularly useful for the separation of gases from a gas mixture by a chemically facilitated separation method such as chemical absorption. An absorbant fluid can be circulated in one membrane channel. The gas mixture is enclosed in a separate channel separated from the absorbant fluid by a semi-permeable membrane (4). The selected absorbed gas component may be removed from the absorbant fluid through a second (7) semi-permeable membrane enclosing a third membrane flow channel in contact with the absorbant fluid.

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# Spirally Wound Membrane Device Having Three Channels

## Background of The Invention

The present invention relates generally to spirally wound membrane devices useful for membrane separations on fluid feed streams. The present invention is broadly useful in the processing of liquids and gases.

## Related Art

Simple, single leaf spirally wound membrane devices have been known for some time. For example, British Patent 489,654 (September 1, 1938) describes a countercurrent spiral membrane device containing a cellulosic membrane material used for dialyzing caustic soda lye. As discussed therein, with such a design it is possible to fit consider ble effective surface area of dialyzing membrane into an extraordinarily small volume, for example, a cylindrical container. Similar single leaf spiral membrane devices are broadly used today in a variety of applications. The advent of reverse osmosis desalination of saline waters in the early 1960's led to a number of improvements in spiral membrane devices, especially in terms of size. U.S. Patent 3,417,870 describes a means of spirally winding a

plurality of reverse osmosis membrane sheets and spacer materials around a perforated hollow core in a manner so as to provide large diameter spiral devices. The design illustrated and described in U.S. Patent 3,417,870 has since been commonly applied to reverse osmosis, ultrafiltration, gas separations, and other types of membrane applications. U.S. patent 4,235,723 describes an alternate method of assembling spiral membrane devices, in which a flow guide member is attached to a perforated hollow core, and a parallel array of membrane envelopes are attached together in a manner such that the permeate from each membrane envelope empties into the flow guide member. Such an arrangement has been termed a "tributary design," since a series of tributaries feed into a main stream, that is, the flow guide member, which empties into the hollow core.

More recently, a number of patents have appeared which incorporate improved materials and modified flow paths in single and multiple leaf spiral elements. U.S. Patent 4,834,881 discloses a variety of improved corrugated spacers for use in feed fluid channels. U.S. Patents 4,8612,487 and 4,902,417 describe improved channel spacers consisting of ribbed plastic nettings. Flow channel spacers in the context of spiral wound membrane elements are occasionally described as two dimensional articles, when the context permits. However, it is essential that the flow channel spacer have a thickness and open volume for the movement of fluid between membrane layers. A similarly ribbed plastic screen useful as a permeate channel flow spacer is described in U.S. Patent 4,476,022. U.S. Patent 3,872,014 describes a design employing a perforated hollow mandrel having a plug located internally so as to

cause a feed stream entering one end to flow out into an attached flow channel leaf. A flow diversion barrier extends out from the hollow mandrel along a fractional length of the attached leaf so as to force the feed stream flow path to encompass the full radial length of the leaf before returning to the hollow mandrel downstream from the plug. Similar approaches are described in U.S. Patents 4,033,878 and 4,765,893. U.S. Patent 4,765,893 also describes the use of a flow diversion barrier extending radially inward from the outer circumference of a spirally wound membrane device on the opposite side of the spirally wrapped membrane, that is, in the flow channel separated from the hollow mandrel compartment by the membrane. Use of a multiple array of flow diversion barriers to achieve a convoluted flow path is described in U.S. Patent 4,814,079 as well as in U.S. Patent 4,033,878.

A commonly shared characteristic of all of these spiral designs is that only two compartments exist therein, each of which is in fluid communication with at least one port. In the case of reverse osmosis, one compartment would consist essentially of a flow channel for the saline feed stream, and the other compartment would consist of a flow channel for the membrane permeate. In the case of pervaporation, one compartment would similarly consist essentially of a flow channel for a liquid feed stream, and the other compartment would consist of a flow channel for permeated vapor removed through the pervaporation membrane. In the case of gas separation, a gas mixture would be fed through one compartment, and a selectively permeated fraction removed through the other compartment. All such spiral designs are capable, therefore, of effecting only a

single separation. If two distinct membrane operations were needed, two separate membrane devices would be required.

In some cases, even a single membrane separation cannot be accomplished satisfactorily in a conventional two-compartment membrane device. Facilitated transport membrane separation is an example of such a case. In facilitated transport, separations between two fluid streams are effected by means of a liquid extractant medium held within the interstices of a porous membrane, the medium being immiscible with the fluid streams on either side. The life expectancy of a facilitated transport membrane is relatively short because of problems associated with loss of the extractant medium and with emulsification and breakthrough. One approach used to solve this problem has been to incorporate two sets of hollow fibers into a hollow fiber membrane module, one for a feed stream and one for a permeate stream, both sets of fibers being immersed in a liquid facilitated transport medium. Such a device is described in U.S. Patent 4,750,918. With such a hollow fiber design, the facilitated transport medium can be separately replenished or renovated by means of ports connected thereto. This application is one of potentially many examples wherein current spirally wound membrane modules are inadequate due to their dual compartment design limitations. The present invention described herein below overcomes the above limitations inherent in dual compartment spiral element designs.

Summary of The Invention

The present invention comprises a spirally wound membrane module having three separate, distinct flow channels for fluid streams. This invention differs from all earlier spirally wound membrane devices by virtue of its triple channel design. The triple channel design bestows a high level of versatility not possessed by earlier spiral designs. Thus, more than one membrane type may be assembled into the triple channel spiral module, and up to three separate fluids may be flowed through the respective channels simultaneously. Each channel may be individually designed to have a combination of inlet and outlet ports; alternatively, one or two of the three channels may be designed to have only outlet ports. Specific combinations of choice on channel flow and porting will depend appropriately upon the desired application or end use.

The triple channel spiral module, in its simplest embodiment, is assembled by winding a serially arrayed pair of membranes upon a perforated hollow mandrel. Such assembly contrasts with other conventional spiral modules where the membranes are always positioned in a parallel array one to another. In the fabrication of a spirally wound membrane module, a membrane sheet is typically folded once upon itself, and the edge delineated by the folding is inserted into a winding nip adjacent to the core, that is, adjacent to the hollow perforated mandrel. In the triple channel spiral module, a second membrane sheet, also typically folded upon itself, is nested within the first folded membrane sheet, to provide a serially positioned membrane set. Appropriate fluid flow channel spacers are located between the membrane layers so as to

delineate flow channels. This serial membrane set is wound upon the core. The three channels are isolated from one another by bonding methods, such as glue application, at the sides and ends of channels where appropriate. A plurality of nested membrane envelopes may be stacked upon one another and wound upon the core, following in a generally analogous manner the approaches used in two-compartment spirally wound membrane modules.

Certain separations, for example those in the pharmaceutical industry require separation of compounds in a molecular weight range from a liquid fermentation broth. The prior art would permit a feed stream to be divided into two streams, a permeate (the portion passing through the semi-permeable membrane) and a retainate (the portion not passing through the semi-permeable membrane). The present method using a triple channel spiral wound membrane permits separations of the feed stream into three streams: the retainate, as with the prior art, a first permeate stream which has passed through one membrane envelope, but not a through a second membrane, and a second permeate stream which has passed through both membrane envelopes.

Still other separations make use of chemical reactions and chemical changes to facilitate a separation. For example, in separating ethylene from ethane in a mixture to the two gases, it is known that ethylene may be selectively absorbed in an aqueous solution of a silver salt. A gas/liquid contactor is necessary for the absorbtion step. Next, a stripper or other means of desorbing the ethylene from the aqueous silver salt solution is required. The present apparatus and method permits a single apparatus to absorb a fluid

in an appropriate facilitating liquid and desorb the fluid from the absorbing liquid.

The inventive triple channel spiral wound membrane element may be comprised of a hollow mandrel having at least one aperture in the wall thereof, a first fluid channel spacer, a portion of which is in contact with said mandrel along an outer surface portion of the mandrel's wall wherein the aperture is located, a first sheet-like membrane having a first and second surface, the first surface being substantially in co-extensive contact with both surfaces of said first fluid channel spacer, the first sheet-like membrane being sealed such that fluid communication with the first fluid channel space is substantially limited to the mandrel bore or through the first sheet-like membrane, a second fluid channel spacer, the second surface of the first sheet-like membrane juxtaposed with a first surface of the second fluid channel spacer, a second sheet-like membrane having a first and a second surface, the first surface being juxtaposed with the second surface of the second fluid channel spacer, a substantially fluid tight seal joining the the second surface of the first sheet-like membrane with the first surface of the second sheet-like membrane at a location radially remote from the mandrel, effectively parallel with the mandrel, such that fluid communication with the second fluid channel space is substantially limited to flow substantially parallel to the mandrel or through the first sheet-like membrane with the first channel space or through the second sheet-like membrane, a third fluid channel spacer, the second surface of the second sheet-like membrane being juxtaposed with both surfaces of the third fluid channel spacer and sealed at the

spiral wound edges such that fluid communication with the third fluid channel space is substantially limited to the unsealed portion at the radially outward extremity of the second sheet-like membrane or through the second sheet-like membrane from the second fluid channel space.

The inventive triple channel spiral wound membrane element may also be described as a first membrane envelope having a fluid flow channel within the membrane envelope in fluid communication with at least one first-fluid port through the containment vessel, a second membrane envelope having a fluid flow channel within the second membrane envelope in fluid communication with at least one second-fluid port, and a third membrane envelope having a fluid flow channel within the third membrane envelope in fluid communication with at least one third-fluid port.

The process of the present invention for separation of fluid components comprises contacting the fluid in a first fluid flow channel enclosed in a first semi-permeable membrane of a spiral wound membrane module, wherein the first membrane has an upper molecular weight cut-off, contacting in a second fluid flow channel the leachate from the first membrane with a second semi-permeable membrane having a lower molecular weight cut-off enclosing a third fluid flow channel, collecting the selected molecular weight fluid fractions from one or more ports in fluid communication with the first fluid flow channel, the second fluid flow channel, and the third fluid flow channel.

The process of the invention as particularly adapted for facilitated transport of a gas component of

a gas mixture comprises contacting a gas mixture in a first fluid flow channel enclosed in a first semi-permeable membrane of a spiral wound membrane module, contacting the first semi-permeable membrane with a chemical fluid which selectively absorbs at least one component of the gas mixture, the chemical fluid is within a second fluid flow channel, contacting the chemical fluid with a second semi-permeable membrane under conditions which cause the at least one component of the gas absorbed in the fluid to desorb and pass through the second semi-permeable membrane into a third fluid flow channel, collecting the desorbed gas from one or more ports in fluid communication with the third fluid flow channel.

The triple channel spiral module design, methods of assembling of such a design, and separations of fluid components advantageously such a design will become evident in the following description of invention.

Brief Description of The Drawings

Figure 1 shows an arrangement of materials useful for winding a triple channel spiral module.

Figure 2 shows a partially fabricated triple channel spiral module.

Figure 3 shows a spirally wound triple channel module complete with containment vessel.

Figure 4 shows a cross-section through the module of Figure 3 at A-A.

Figure 5 shows a cross-section through the module of Figure 3 at B-B.

Figure 6 shows a hollow mandrel having a plug located intermediate in its length, and an attached fluid flow channel spacer having a flow diversion barrier therein.

Figure 7 shows a triple channel spiral module complete with containment vessel wherein each channel has an inlet port and an outlet port.

Figure 8 shows the area identified as 37 in Fig. 2 in an expanded view of a typical seal between the second surfaces of a second membrane sealing a third fluid flow channel along the spiral wound edges while leaving the radially remote edge of the third flow channel open to fluid flow.

Description of a Preferred Embodiment

Figure 1 shows an arrangement of materials which, when spirally wound, give rise to a triple channel membrane device. A layer of a first channel spacer material 1 is attached or otherwise in contact with the outer periphery of a hollow mandrel 2 having at least one aperture 3 therein for fluid communication between the mandrel's interior and external zones. Channel spacer material preferably at least one layer of this first channel spacer material 1 is juxtaposed between any subsequent membrane layers and the outer surface of the mandrel 2, so that all of the outer periphery of the mandrel 2 is in contact with a first flow channel thereby formed. A layer of a first membrane 4 is placed in contact with the first layer of channel spacer 1. The membrane 4 is preferably a sheet-like material. This layer of membrane 4 is conveniently folded upon itself, forming a fold edge 5 that is positioned parallel to the axis of the mandrel 2. First

membrane 4 has a first surface 4a and a second surface 4b. The first surface 4a is in contact with the first spacer material 1. The second surface ·b is in contact with a second channel spacer material 6. This second channel spacer material 6 is interposed between the first membrane 4 and a second membrane 7. The second membrane 7 is preferably folded upon itself just as with the first membrane 4, and is typically nested within the first membrane fold during a spiral winding assembly operation. The second membrane 7 has a first surface 7a and a second surface 7b. The first surface 7a is in contact the second channel spacer material 6. The second surface 7b is in contact with a third channel spacer 8. Thus, three flow channels are formed, the first (or inner) flow channel being effectively separated from the second (or middle) flow channel by the first membrane 4, and the second (middle) flow channel being separated from the third (or outer) flow channel by the second membrane 7, when these layers are spirally wound upon a core along with appropriate glues or bonding agents at the channel edges as will be more fully described hereafter. By the expression glues or bonding agents at the channel edges, the practitioner will understand a benefit results from locating the glue lines near the edge of the spiral wound membrane element and near the radial edge of a membrane/flow channel. A resulting benefit is to make maximum use of the membrane area included in the spiral wound element. Of course less advantageous placement of the glue lines is possible and contemplated.

While Figure 1 and the above description utilize folded sheets of the membranes, two pieces of a membrane may also be bonded or spliced together at an

edge, the spliced or bonded edge serving as an insertion point into the assembly arrangement, just as with the folded edge. Also, the second channel spacer 6 need not be a single folded sheet. It may optionally be a spliced pair of sheets as well, or may even consist of two sheets, one placed above and the other placed below the nested membrane 7.

Turning now to Figure 2, these layers of membrane and channel spacer material are wound upon the mandrel 2, with appropriate application of a bonding material such as a glue so as to isolate the three resulting flow chambers one from another. Figure 2 shows the hollow mandrel 2 having already wrapped about it a layer of the first channel spacer 1 and a layer of the first membrane 4. Both layers together are bonded to the mandrel 2 in zones 9a and 9b by means of a glue, sealant, or other agent. The first membrane 4 extends radially outward from the mandrel 2, enveloping in a typically co-extensive manner on both sides the layer of first channel spacer material 1 that also extends radially outward from the mandrel 2. The two side regions 10a, 10b and the end region 10c of this membrane envelope leaf are sealed by means of a glue, sealant, or other bonding method, thereby creating the first flow channel in the open volumes provided by the first channel spacer material. This first channel is open only to the mandrel, and is in fluid communication with the hollow core of the mandrel by means of at least one perforation or aperture 3 through the wall of the mandrel. The bonding method for sealing the three sides of this flow channel and for sealing the envelope around the mandrel is conveniently accomplished by means of a two-part urethane or epoxy glue which, upon being mixed,

has an appropriate pot life for working and for movement and assembly of the layers of materials spirally around the mandrel before the glue gels and hardens. Other types of bonding may be employed, such as for example, by ultrasonic welding, gluing with hot melt adhesives, or sealing with moisture-activated silicone sealants, although some of these may increase the complexity of the spiral winding assembly operation.

The second channel spacer material 6 is shown in Figure 2 as a sheet folded upon itself, the fold edge being inserted into the nip made by the first membrane 4 at the mandrel 2. This second channel spacer can consist of material identical to the first channel spacer, or may be entirely different. Spacer materials may consist of porous fabrics, plastic nets, corrugated plastic sheeting, metallic screens, or any of a wide variety of materials. In Figure 2, the spacer is depicted as an open netting, but subsequent figures depict it as a corrugated spacer for sake of clarity. Similarly to the folded channel spacer 6, a fold edge of the second membrane 7 is inserted (nested) into the nip thus formed by this second channel spacer material. The folded sheet of this second membrane 7 is shown as being coextensive with a sheet or layer of the third channel spacer material 8, though this is not a strict requirement for membrane 7 (or for membrane 4). The membrane sheet may allowably extend beyond the length of the channel spacer sheet, for example.

The two sides of the membrane envelope formed by side 7b of the second membrane 7 and the third channel spacer material 8 are sealed from fluid flow by a glue 12d or other bonding means in zones 12a, and 12b similar to the first membrane envelope. At the distal

end 12c of this second membrane envelope the third channel spacer and the side 7b of the second membrane 7 is left at least partially unsealed. As a consequence of not sealing the distal edge 12c the channel formed by means of the channel spacer material 8 sandwiched between the folded layers of membrane 7 may be in fluid communication with external ports in a resulting module.

A sealing means 11 such as a bead of a glue is disposed on each of the distal ends of the second channel spacer material 6 effects a leak-free bond between the surface 7a and surface 4b of the first membrane 4, when these layers are spirally wound into a membrane scroll upon the hollow mandrel 2.

Fig. 8 is an enlarged view of a corner of a flow channel formed about channel spacer material 8 by membrane sheet 7 and glue or bonding agent 12d. The unsealed end 12c of the membrane envelope is readily seen to be open to fluid communication with external fluid ports of a resulting membrane module. A membrane module identifies a spiral wound membrane element adapted for fluid connections within a containment vessel.

Further clarification of the design of this triple channel spiral element may be gained by reference to Figures 3, 4, and 5. Figure 3 depicts a triple channel spirally wound membrane module complete with a containment vessel 13 for fluid treatment. Two sets of nested membrane envelopes are present in the membrane scroll of this module, which will become evident later by reference to the A-A cross-section depicted in Figure 4. The spirally wound membrane element is contained in a vessel 13 having one or more ports 14 for flow

associated with the channel formed by the first channel spacer material 1, one or more ports 15 for flow associated with the channel formed by the second channel spacer material 6, and one or more ports 16 for flow associated with the channel formed by the third channel spacer material 8. The membrane and spacer layers have been spirally wound upon a hollow mandrel 2, which is then brought into connection with its ports 14. Extensions 22 of the mandrel 2 beyond the ends 23 of the membrane scroll can themselves serve as ports through the ends of the containment vessel, as depicted in Figure 3. A seal to the mandrel extension 22 can be made by means of an O-ring 19 mounted in an orifice within an end plate 20 held in position by a snap ring 21. This example is an illustration of just one of several means known in the art for porting the fluid stream flowing from the internal space of the hollow mandrel 2. The external periphery of the spirally wound element is fitted circumferentially with sealing cuffs 17 at each end, which are in a sealing contact with the inner wall of the vessel 13. These cuffs 17 in combination with the sealant means 11 (see Figure 2) effectively isolate the second channel ports 15 from the third channel ports 16. One leaf ending 18 is shown in Fig. 3. The c responding leaf ending for the other third channel [illegible] not shown in this figure, since it would customarily have its terminus at 120 to 180° from the first ending 18. The open ended channel 12c of Figure 2 comprises the open gap between the leaf ending 18 and the underlying membrane surface, with the third channel spacer 8 shown in Figure 2 herein extending outward from the open end and wrapping once around the outer circumference.

Figure 4 shows a cross-section at A-A for this spirally wound device, illustrating the cross-section of the double set of nested membrane envelopes. Herein, the terminus 24 of the secondary membrane envelope (i.e., the envelope formed by the second membrane 7 and the third channel spacer material 8) would correspond to the leaf ending 18 in Figure 3, and the terminus 25 of the companion secondary membrane envelope is shown as being on the opposite side of the device. The cross-section in Figure 4 also shows: a first flow channel 26 associated with the first channel spacer material 1, which is in fluid communication with the perforated mandrel 2; a second flow channel 27 formed by the second channel spacer material 6, which is in fluid communication with the enclosed volume 34 formed by the vessel 13 and the end of the membrane scroll 23; and a third flow channel 28, which is associated with the third channel spacer material 8. The third flow channel 28 opens into, and is in fluid communication with, an annular space 29 between the membrane scroll and the internal surface 30a of the vessel wall 30, and is in fluid communication with a port 16. The folded edges of the first membranes (corresponding to 4 in Figs 1 and 2 are shown at their insertion points 31a, and 31b). Channel endings of the flow channels 26 enclosed by these first membranes 4 are shown at terminus points 32a, 32b. The folded edges of the second membranes (corresponding to 7 in Figs 1, 2, and 3) are shown at their insertion points 33a, 33b, and the corresponding terminus points at 24 and 25. The third channel spacer 8 is preferably and conveniently extended beyond the terminus points 24 and 25, being wrapped once, or more, around the outer circumference of the membrane scroll so

as to enhance fluid flow between channel 28 and the fluid port 16.

It will be evident to practitioners skilled in the art that the various layers of membranes and channel spacers need not end exactly at the leaf terminus points; individual layers may very well extend beyond the terminus points without altering the functional features shown in Figs. 3 and 4.

Figure 5 shows a cross-section at B-B. The sealing relationship of the cuff 17 between the outer circumference of the spirally wound element and the inner surface 30a of the containment vessel wall 30 is illustrated in this figure. Essentially, the cuff is formed by filling the annular space 29 with a sealing means to block annular flow or mixing between channels 27 and 28 at the spiral wound element periphery. Also at section B-B is a glue or sealant in flow channels 26 and 28 at their exposed ends at the edge 23 of the membrane scroll, corresponding to zones 10a, 10b and 12a, 12b in Figure 1. Flow channel 27 is open, however, and flow in a vector orthogonal to the plane of cross-section B-B is permitted. The hollow mandrel 2 need not have provision for wall apertures in this region, of course. The cuff 17 may consist of a bead of sealant, such as derived from a silicone, urethane, or epoxy resin composition. It may also consist of an annular rubber ring or shaped rubber seal. Alternatively, the cuff may consist of a molded or machined annular sleeve of plastic or metallic composition which fits over the outer circumference of the wound membrane scroll, and contains a channel for a rubber O-ring or gasket on its outer periphery, so that a seal can be made to the inner wall of the containment vessel. Open spaces between

such plastic or metallic sleeve and the membrane scroll can be filled with a sealant. A variety of such techniques are known in the art of spirally wound membrane elements. A potential advantage associated with annular sleeves of plastic or metallic composition is that their use would fix the outer dimension of spirally wound membrane scrolls, preventing them from unwinding. Wrapping the outer periphery of membrane scrolls such as with a pressure sensitive adhesive tape may also be employed to maintain the scroll in its wound condition, as long as some openings are provided in the tape wrap for interchange of fluid between the third channel 28 and the annular space 29.

Each of the three flow channels can be designed to accommodate both an inlet flow into and through the channel to an exit port. Figure 6 shows a modified hollow mandrel 40, coupled with a modified flow channel spacer sheet 41 which, upon substitution into the arrangement shown in Figure 2, will lead to a triple channel spiral module having a flow-through capability in the first channel 26 (Figure 4). In Figure 6, the hollow mandrel 40 has a plug 42 in its hollow core at some point generally midway in its length. Apertures 3 are located in the walls of the mandrel 40 on both sides of the plug. A first permeate channel spacer material 1 is attached to the mandrel 40, and extends radially outward therefrom. A flow diverting means 43 is positioned generally near or on the midline of the sheet of permeate channel spacer material 1, extending radially outward from the mandrel 40. The flow diverting means 43 does not extend to the distal end of the spacer material sheet; however, provision is made for fluid flow to pass from the first side of the sheet

around the tip of the flow diverting means 43 to the second side of the sheet. Thus, a fluid can be introduced through one of the modified mandrel 40, exit through apertures 3 into the flow channel defined by the channel spacer material 1, percolate down to the distal end of the flow channel on one side of the sheet, return to the core on the other side of the sheet, re-enter the hollow mandrel 40 by apertures 3 and exit the mandrel 40 through an appropriate port. The outer boundaries of the flow channel can be fixed by means of glue lines or other sealing methods in zones 10a, b, and c as described earlier. Means for diverting the flow may include, for example, a band of a glue or sealant, a foam strip, or a strip of a plastic or metal. It need not be a perfectly impervious barrier to achieve the intended effect. The flow diverting means 43, shown in Fig. 6, can be eliminated while still maintaining an inlet-to-outlet fluid flow in the flow channel. However, this would generally not be preferable because it would allow inlet flow to selectively channel to the outlet by the path of least flow resistance. Other shapes and designs for flow diversion through a channel spacer sheet are possible and include those described in U.S. Patent 5,034,126, and U.S. Patent Application 385,230 allowed allowed July 19, 1991.

In a manner analogous to the approach in Figure 6, it is possible to apply a flow diverting means to the third channel spacer sheet 8, and, by means of an inlet and an outlet port, establish capability for inlet-to-outlet flow in the third channel 28. In such a case, the flow diverting means would extend from the outer terminus 18 radially inward towards, but not all the way to, the insertion point of the third channel spacer

material 8 within the folded second membrane 7. The flow diverting means would preferably be mated at the membrane scroll's outer periphery with an additional sealing cuff so as to separate inlet and outlet zones of the annular space 29 corresponding to inlet and outlet ports.

Figure 7 illustrates a membrane module comprising a triple channel membrane element and a containment vessel having provision for inlet-to-outlet fluid flow for all three channels of a triple channel device. The spirally wound membrane device is enclosed in a containment vessel 13 having an endplate 20 at each end, held in place by a restraining means such as a snap ring 21. An inlet port 45 is connected to a modified hollow mandrel 40 (indicated by a dashed line) by means of a coupling 46. The mandrel modification may be a plug 42 as illustrated by Fig 6, or other means to accommodate a fluid inlet and a fluid outlet through the mandrel for a first flow channel. The outflow end of this mandrel 40 may be similarly connected to an outlet port 47. Inlet 48 and outlet 49 ports for fluid flow through a second fluid flow channel formed by a second channel spacer are positioned in the wall 30 of the vessel. These two ports may alternatively be located in the opposing endplates 20 of the vessel. Cuffs 17 at each end of the spirally wound element seal off the flow chamber for the second fluid flow channel from the annular space 29. A cuff 50 is positioned at a location intermediate to the end cuffs on the spiral element periphery. The intermediate cuff 50 divides the annular space 29 into a first annular compartment 29a and a second annular compartment 29b. The first annular compartment 29a is in fluid communication with an inlet

po· 51, the second annular compartment 29b is in fluid communication with an outlet port 52. Fluid entering the inlet port 51 into the annular compartment 29a would thereupon enter into the flow channel defined by the third channel spacer material at the exposed leaf terminus 18a. The fluid would typically be guided by a flow diverting means within the fluid flow channel. After circulating through the fluid flow channel, the fluid exits from the third fluid flow channel at the terminus zone 18b into annular space 29b, and therefrom passing through exit port 52.

The device illustrated in Figure 7 is representative of only one means by which the concept of the invention may be employed. Other variations within the context of this invention may be readily apparent to the practitioner skilled in the art. Inlet-to-outlet flow may be employed in one, two, or all three channels. If inlet-to-outlet flow were employed in only one of the channels, each of the other two channels would naturally function essentially as outflow channels, producing for example a filtrate or permeate stream by filtration or permeation of fluid through the membrane layers. In this particular example, inlet-to-outlet flow would preferably take place through the middle channel. Utility may even be found where no inlet-to-outlet flow occurs in any of the three channels. Rather, a dead end filtration design may be employed, such as where a fluid to be filtered through a microfiltration membrane enters into the middle channel, and filtrate exits from the first and third channels.

Inlet-to-outlet flows are arranged in Figure 7 to provide generally counter-current or cross-current flow of fluid streams along the surfaces of the

membranes contained in the device, but the invention is not limited by this flow arrangement. Co-current, cross-current and counter-current flow paths are all considered within the overall context of the invention, as well as reversal of flow directions during operation of the device.

The two membranes which define, in conjunction with the three channel spacer layers, the three channels of the triple channel spiral device need not be identical one membrane to the other. The two membranes may differ from one another in composition and performance function. Thus, one membrane could be a polyamide-based composition performing as reverse osmosis membrane, for example, the other being a silicone-based composition performing as a pervaporation membrane. Thus, potential exists for simultaneously performing two different types of membrane-based separations on a fluid feed stream. Membranes can be chosen to provide separations in the areas of reverse osmosis, nanofiltration, ultrafiltration, microfiltration, dialysis, pervaporation, coupled transport separations, gas separations, piezodialysis, membrane distillation, solvent vapor recovery, and so forth. Any number of nested membrane sets may be wound into spiral modules of this triple channel design to achieve a variety of module sizes and membrane areas. Individual single sheets of the two membranes may also be accordion-folded (i.e., multiply pleated), with appropriate interleaving of channel spacer materials, to delineate a plurality of membrane envelopes wound upon the mandrel, wherein the first membrane layer may consist of a continuous single sheet of membrane, as may the second membrane layer also. The overall area of the

first membrane in a triple channel device may also be varied considerably relative to the overall area of the second membrane, depending upon the intended application of the device. Other aspects and modifications of this novel triple channel design will become apparent to the practitioner skilled in the art.

Semi-permeable membrane suitable for use in the inventive process and apparatus may be selected from materials exhibiting appropriate chemical and physical properties. Microporous polyethylene, polypropylene, and polytetraflouroethylene membranes may be selected for hydrophobic properties. Such hydrophobic membranes would find advantage in a chemically facilitated gas separation where the facilitating fluid is aqueous based as the membrane sheet would not be wetted by the aqueous based fluid.

Polyamide, and composite polyamide membranes may be selected for hydrophilic properties. Such hydrophilic membranes would find advantage in a separation of molecular weight fractions from an aqueous based solution such as a fermentation solution where wetting of the membrane sheet would be advantageous.

The process of the invention is particularly attractive for separation of feed-gas mixtures utilizing a chemical facilitation means. A feed comprising a mixture of gases is circulated to a first envelope consisting of a semi-permeable membrane. The membrane is permeable to at least one gas component of the mixture. In contact with the first semi-permeable membrane envelope is a fluid which electively facilitates, as by absorption, the removal of at least one gas component from the mixture. The facilitating

fluid is itself contained within a second membrane envelope. A third envelope formed of a membrane semi-permeable to at least one component absorbed by the facilitating liquid is also in contact with the facilitating liquid. At least one component absorbed in the facilitating liquid is stripped from the facilitating liquid.

The facilitating liquid used in the process will in general be selected for the particular gas mixture separation attempted. For instance, facilitating liquid for carbon dioxide as a permeate gas is preferably an aqueous solution of a alkali-metal carbonate. Alternative facilitating liquids include monoethanolamine, diethanolamine and hindered amines, as aqueous solutions or undiluted. Carbon monoxide may be absorbed from a mixture with carbon dioxide by an aqueous solution of cuprous chloride and an alkali-metal chloride such as potassium chloride. Ethylene and propylene may be selectively absorbed from a gas mixture including ethane and propane in an aqueous solution of silver nitrate or other water-soluble silver salt. Hydrogen sulfide may be selectively absorbed from natural gas by an aqueous solution of an alkali-metal carbonate such as potassium carbonate. Sulfur dioxide may be selectively absorbed from stack gas by an aqueous solution of an alkali metalbisulfite or sulfite or polar organic compounds such a sulfolane or polyethylene glycol.

Variations and modifications of the apparatus and methods described will be apparent to the practitioner skilled in the art of fluid separations by semi-permeable membranes. All such variations and

modifications are considered within the scope of the claims below.

Examples

Example 1

A 50 liter sample of a biological fermentation liquor containing a range of molecular weight compounds up to 20,000 including growth nutrients, growth products, cell fractions and inorganic salts is circulated through inlet and outlet ports of a triple channel spiral wound membrane module at (400 psig). The membrane enclosing the inlet fluid flow channel has a molecular weight cut off of 10,000. The second membrane of the triple channel membrane element has a molecular weight cut off of 8,000. Fluid from the second membrane envelope in contact with both the first and second membranes is recirculated through inlet and outlet ports of the second membrane envelope with a pressure maintained at (350 psig). Makeup water of 50 liters is added to the initial sample sufficient to maintain a recirculating sample fluid level of 20 liters.

When 80 liters of fluid has passed through the membrane having a molecular weight cut off of 10,000, 5 liters of a fraction of biological fermentation fluid having a molecular weight range from 8000 to 10,000 has been collected as the recirculating fluid from the second fluid flow envelope.

Example 2

A triple channel spiral wound membrane module substantially configured according to Fig. 7, except as noted, is used to separate a gas stream containing ethylene and ethane in a volume ratio of 1:2 by means of

a chemically facilitated transport. The mixed gas stream is circulated to a first annular compartment corresponding to 29a in fluid communication with a microporous semi-permeable hydrophobic third membrane envelope such as a polyethylene having a porosity of 0.3 and a pore size range from 0.05 to 2.0 µm. An exhaust of the mixed gas stream is removed from the second annular compartment corresponding to 29b. A third channel spacer material is provided with a flow diverting means comprising a line of polyurethane glue.

An aqueous solution of silver nitrate is circulated through a second membrane envelope at a rate of 7.6 l/min (2 gpm) through an inlet port corresponding to 48 and an outlet port corresponding to 49.

A first membrane envelope also comprises of microporous polyethylene. The first channel spacer material has no flow diverting means. The mandrel does not contain a plug. A valve connected the mandrel inlet port is closed. A vacuum is connected to the mandrel outlet port.

The feed gas to the third membrane envelope beginning at a pressure of 103 kPa (15 psig) the pressure of the circulating feed gas becomes 69 kPa (10 psig) after circulation of 10 minutes. Thereafter gas circulation continued. There no significant reduction of pressure in the feed gas after the feed gas stream is circulated for 20 minutes. The ethylene is quickly and efficiently separated leaving the feed gas comprising essentially ethane.

WHAT IS CLAIMED IS:

1. A triple channel spirally wound membrane element, comprising:

a) a hollow mandrel having at least one aperture in the wall thereof;

b) a first fluid channel spacer, a portion of which is in contact with said mandrel along an outer surface portion of the mandrel's wall wherein the aperture is located;

c) a first sheet-like membrane having a first and second surface, the first surface being substantially in co-extensive contact with both surfaces of said first fluid channel spacer, the first sheet-like membrane being sealed such that fluid communication with the first fluid channel space is substantially limited to the mandrel bore or through the first sheet-like membrane;

d) a second fluid channel spacer;

e) the second surface of the first sheet-like membrane juxtaposed with a first surface of the second fluid channel spacer;

f) a second sheet-like membrane having a first and second surface, the first surface being juxtaposed with the second surface of the second fluid channel spacer;

g) a substantially fluid tight seal joining the the second surface of the first sheet-like membrane with the first surface of the second sheet-like membrane at a location radially remote from the mandrel, effectively parallel with the mandrel, such that fluid communication with the second fluid channel space is substantially limited to flow substantially parallel to the mandrel or through the first sheet-like membrane with the first channel space or through the second sheet-like membrane;

h) a third fluid channel spacer;

i) the second surface of the second sheet-like membrane being juxtaposed with both surfaces of the third fluid channel spacer and sealed at the spiral wound edges such that fluid communication with the third fluid channel space is substantially limited to the unsealed portion at the radially outward extremity of the second sheet-like membrane or through the second sheet-like membrane from the second fluid channel space.

2. A membrane module comprising a triple channel spirally wound membrane element of Claim 1, and a containment vessel having a communication port in fluid communication with each fluid channel space.

3. A membrane module according to Claim 2 wherein at least one fluid channel spacer is in fluid communication with more than one fluid communication port.

4. A spirally wound membrane module comprising a containment vessel, and a membrane element comprising:

a) a first membrane envelope having a fluid flow channel within the membrane envelope in fluid communication with at least one first-fluid port through the containment vessel,

b) a second membrane envelope having a fluid flow channel within the second membrane envelope in fluid communication with at least one second-fluid port, and

c) a third membrane envelope having a fluid flow channel within the third membrane envelope in fluid communication with at least one third-fluid port.

5. The device of Claim 4 wherein the membrane envelopes of the first and third membrane envelopes have the same chemical composition or structure.

6. The device of Claim 4 wherein the membrane envelopes of the first membrane envelope has a chemical composition or structure different from that of the second membrane envelope.

7. The device of Claim 4 wherein the fluid flow channel of at least one of the membrane envelopes is in fluid communication with both an inlet port and an outlet port.

8. The device of Claim 4 wherein means for diverting flow is provided on a fluid channel spacer within at least one membrane envelope.

9. The device of Claim 1 wherein means for diverting flow is provided on at least one fluid channel spacer.

10. A method of separating a fluid containing component having a range of molecular weights comprising:

contacting the fluid in a first fluid flow channel enclosed in a first semi-permeable membrane of a spiral wound membrane module, wherein the first membrane has an upper molecular weight cut-off,

contacting in a second fluid flow channel the leachate from the first membrane with a second semi-permeable membrane having a lower molecular weight cut-off enclosing a third fluid flow channel,

collecting the selected molecular weight fluid fractions from one or more ports in fluid communication with the first fluid flow channel, the second fluid flow channel, and the third fluid flow channel.

11. A method of separating a mixture of gases comprising:

contacting a gas mixture in a first fluid flow channel enclosed in a first semi-permeable membrane of a spiral wound membrane module,

contacting the first semi-permeable membrane with a chemical fluid which selectively absorbs at least one component of the gas mixture, the chemical fluid is within a second fluid flow channel,

contacting the chemical fluid with a second semi-permeable membrane under conditions which cause the at least one component of the gas absorbed in the fluid to desorb and pass through the second semi-permeable membrane into a third fluid flow channel,

collecting the desorbed gas from one or more ports in fluid communication with the third fluid flow channel.

12. The method of Claim 11 wherein the gas mixture comprises ethylenically unsaturated hydrocarbons saturated hydrocarbons of 6 carbon atoms or less.

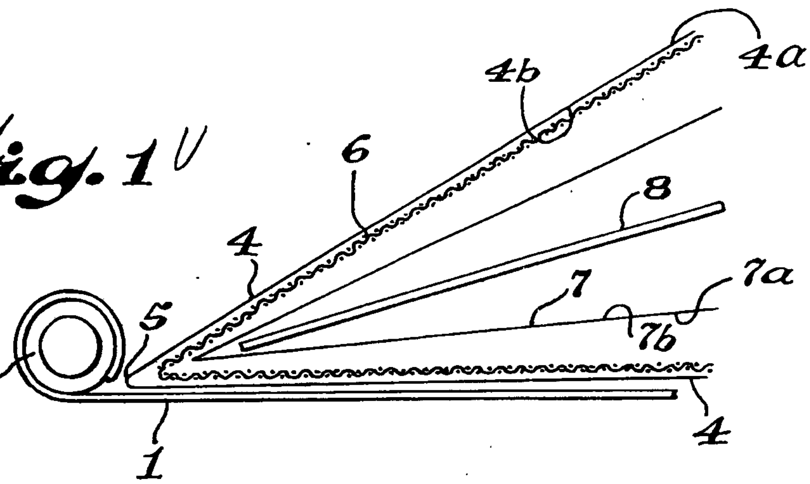
13. The method of Claim 12 where the gas mixture comprises ethylene/ethane, propylene/propane, butene/butane, or pentene/pentane mixtures.

14. The method of Claim 11 where the gas mixture comprises methane/nitrogen, methane/helium, sulfur oxides/air, nitrogen oxides/air, carbon dioxide/methane, or hydrogen sulfide/hydrocarbons.

15. The method of Claim 11 where the chemical fluid is a aqueous solution of a silver salt, a copper salt, an alkali-metal chloride, an amine, alkali-metal carbonate, alkali metal bisulfite.

16. The method of Claim 11 where the chemical fluid is a polar organic compound, or a neat amine.

Fig. 1



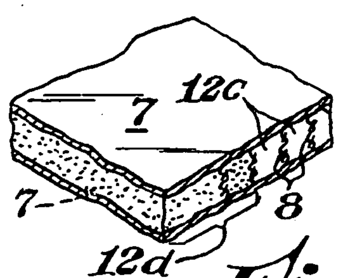




Fig. 8

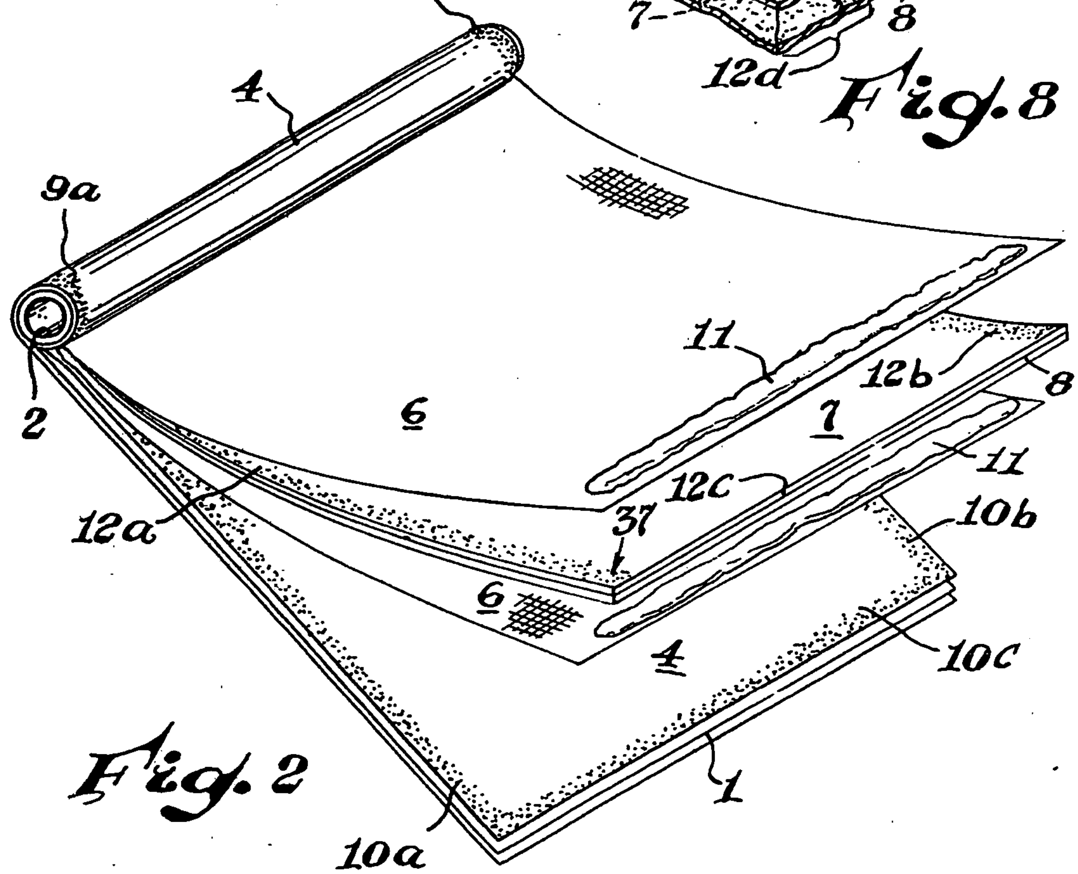



Fig. 2

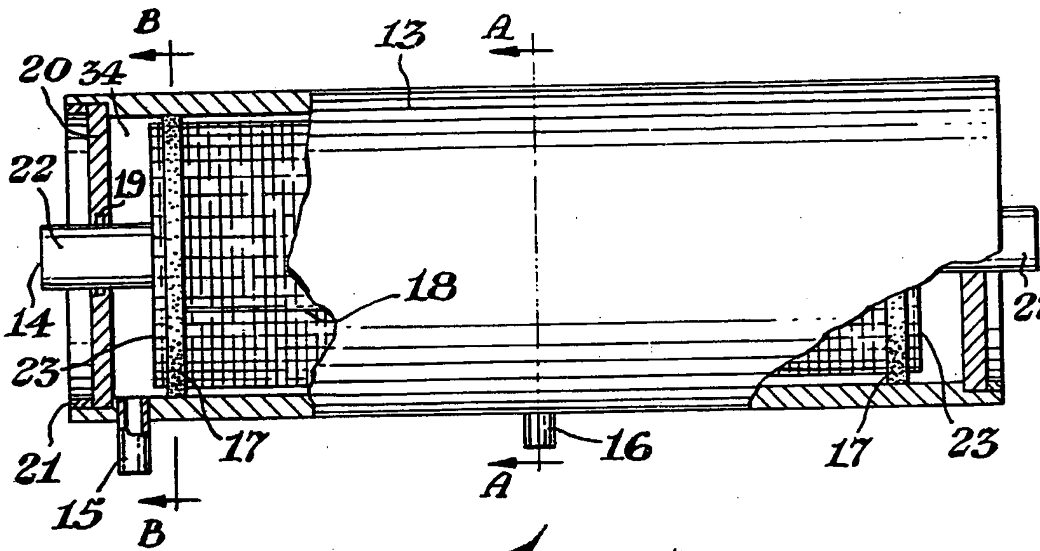

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Fig. 3

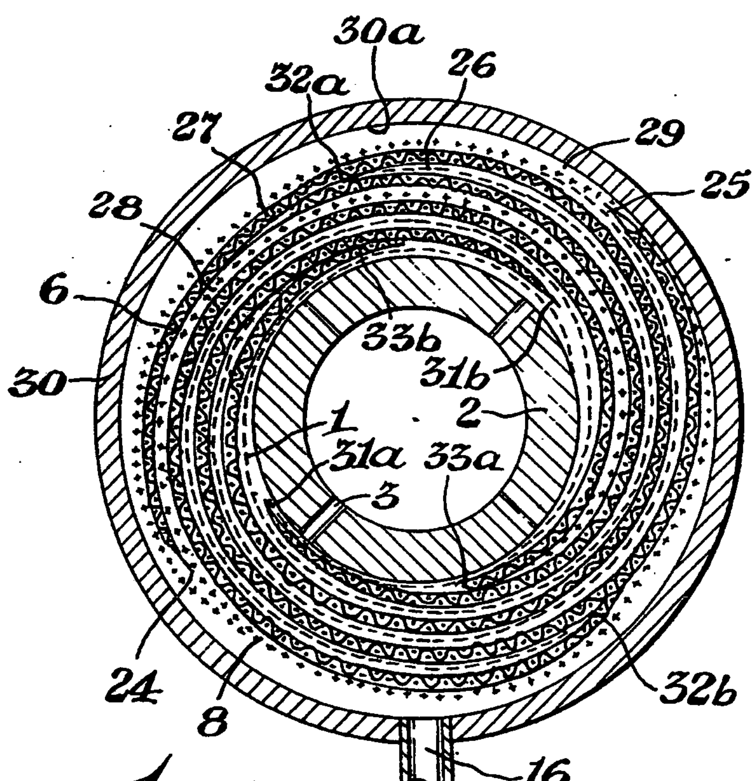
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Fig. 4

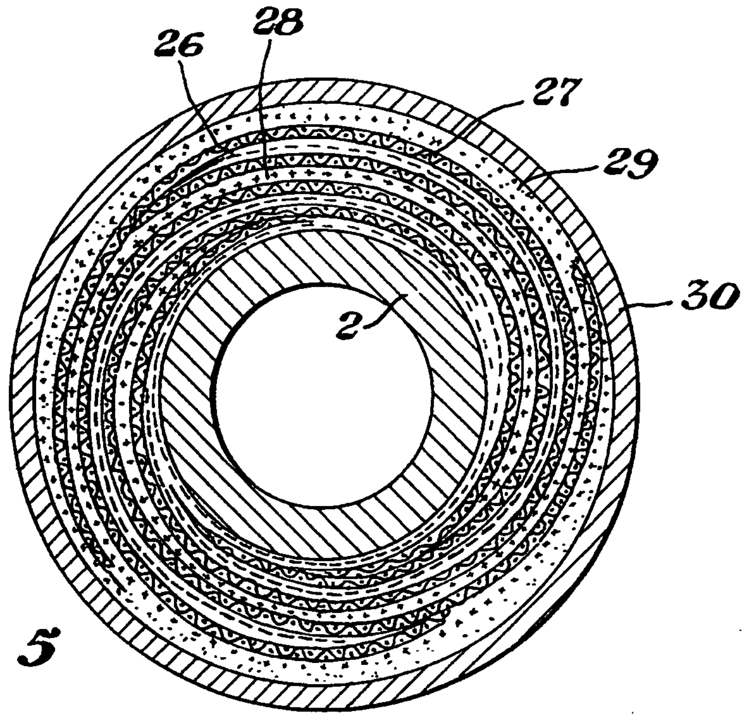


Fig. 5

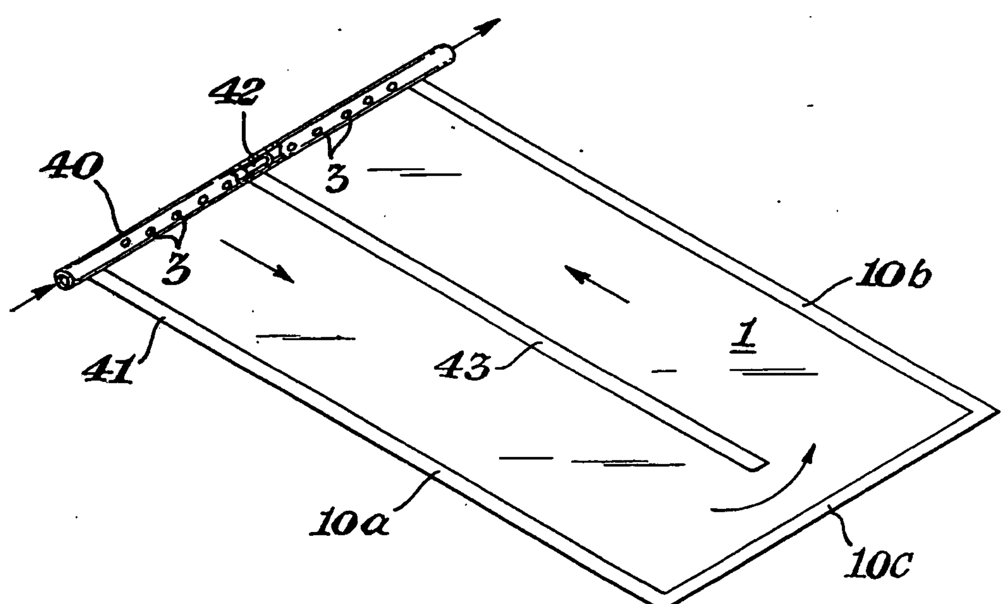


Fig. 6

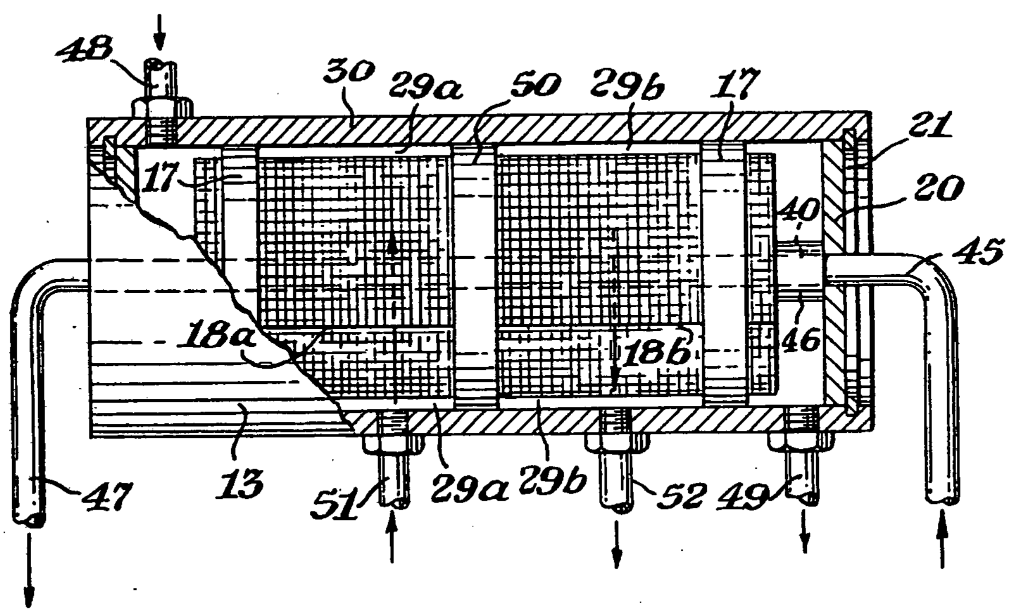


Fig. 7

# INTERNATIONAL SEARCH REPORT

International Application No. PCT/US91/08825

**I. CLASSIFICATION OF SUBJECT MATTER** (if several classification symbols apply, indicate all) [6]

According to International Patent Classification (IPC) or to both National Classification and IPC

IPC(5) B01D 61/28
U.S. CL. 210/638

**II. FIELDS SEARCHED**

Minimum Documentation Searched [7]

| Classification System | Classification Symbols |
|---|---|
| U.S. | 55/16.158<br>210/638,641,644,321.74,34.83, 494.1,487 |

Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched [8]

**III. DOCUMENTS CONSIDERED TO BE RELEVANT** [9]

| Category * | Citation of Document, [11] with indication, where appropriate, of the relevant passages [12] | Relevant to Claim No. [13] |
|---|---|---|
| A | US,A, 3,872,014 PUBLISHED 18 MARCH 1975, SCHELL | 1-16 |
| A | US,A, 5,034,126 PUBLISHED 23 JULY 1991 REDDY et al. | 1-16 |

* Special categories of cited documents: [10]

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

**IV. CERTIFICATION**

| Date of the Actual Completion of the International Search | Date of Mailing of this International Search Report |
|---|---|
| 04 FEBRUARY 1992 | 28 FEB 1992 |
| **International Searching Authority** | **Signature of Authorized Officer** |
| ISA/US | Ngoc-Ho Nguyen<br>for FRANK SPEAR NGUYEN NGOC-HO<br>INTERNATIONAL DIVISION |